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# Investigation of the Characteristics of Ferroelectric Thin Films Deposited By Pulsed Laser Ablation

S. Sengupta, N. Sonnenberg, D.P. Vijay, and S.B. Desu

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## INTRODUCTION

A ceramic Barium Strontium Titanate,  $\text{Ba}_{1-x}\text{Sr}_x\text{TiO}_3$ , phase shifter using a planar microstrip construction has been demonstrated.<sup>1</sup> In order to obtain higher operating frequencies (beyond 30 GHz) and to decrease the voltage requirements, thin film fabrication of the above candidate materials is necessary. The electronic characteristics of BSTO and oxide-modified BSTO films have already been demonstrated.<sup>2,3</sup>

In this work, we have investigated the effect of the thin film stoichiometry on electrical performance by measuring the dielectric constants and tunability in the low frequency (KHz) range. A HP 4194A impedance analyzer was used for these measurements. The results of these measurements will be discussed.

## EXPERIMENTAL

The lattice parameters and dielectric constants of the substrates used in these experiments are listed in Table I. Prior to Pulsed Laser Deposition (PLD), the substrates underwent a cleaning process that included an ultrasonic cycle in TCE followed by two methanol ultrasonic cycles. The samples were then rinsed with methanol and air dried.

TABLE I. Lattice Parameters and Dielectric Constants for  $\text{MgO}$ ,  $\text{Al}_2\text{O}_3$  and  $\text{LaAlO}_3$  Substrates.

	<u>SUBSTRATE</u>		
	<u>MgO</u>	<u>Al<sub>2</sub>O<sub>3</sub></u>	<u>LaAlO<sub>3</sub></u>
Lattice Parameter (Å)	4.21	4.76	3.79
Dielectric Constant (300K)	10	11	23

The ceramic ablation target chosen for this work was  $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3$  (BSTO). In order to measure the electronic properties of the films, a ground plane electrode of Ruthenium oxide ( $\text{RuO}_2$ ) was sputtered onto the substrates at a substrate temperature of 200 °C and a  $\text{O}_2/\text{Ar}$  ratio of 1:4 with a total pressure of 10 mT. The Ruthenium oxide films were 3000 Å thick. The thickness measurement was performed using a Dektak-200 profilometer. The resistivity of the as-deposited films was on the order of 160  $\mu\text{ohms-cm}$ . These were annealed at 600 °C for 30 minutes to lower the resistivity and then cooled by furnace quenching. The resistivity of the annealed films was measured to be 110  $\mu\text{ohms-cm}$ .

The PLD was accomplished using a Questek 2000 krypton-fluoride excimer laser with a wavelength of 248 nm and a repetition rate of 10 Hz. The substrate was parallel to the target with a separation distance of 55 mm. The average pulse energy was 300 mJ with a 20 ns pulse width. The substrate temperature was 500 °C, as monitored by a thermocouple clamped between the heater and the substrate. The powder pressed ceramic targets were prepared according to a

description published previously.<sup>2</sup> The oxygen partial pressures used for this experiment were 150 mT or 50 mT. A Dektak-200 profilometer was used to measure the film thickness that was approximately 6000 Å on all the substrates.

The thicknesses of the top Pt electrodes were measured to be approximately 3000 Å also using profilometry. The metallized films used for the electrical measurements consisted of Sapphire/RuO<sub>2</sub>/BSTO / Pt. Thin films of BSTO were also deposited on bare substrates of MgO and LaAlO<sub>3</sub>.

The glancing angle x-ray measurements were performed using a Rigaku RU 200 rotating anode x-ray diffractometer. The entrance slit had a width of 0.2° and the angle of incidence of the Cu Kα ( $\lambda=1.5415$  Å) beam was set at 1°. The step scanning interval was 0.02° and the scan rate was 5°/minute.

The dielectric constant ( $\epsilon'$ ) and % tunability were determined for all thin film/substrate combination. The % tunability of a material is determined using the following equation:

$$\% \text{ tunability} = \{ \epsilon'(0) - \epsilon'(V_{app}) \} / \{ \epsilon'(0) \} \quad (1)$$

The tunability measurements were taken with an applied electric field which ranged from 0 to +/- 2.0 V/micron ( $\mu\text{m}$ ). The electronic properties were measured using a 30 KHz frequency. Capacitance versus voltage (C-V) measurements for the films were taken using an HP4194 impedance / phase gain analyzer. The voltage, applied internally through the HP 4194A, was varied from -1.2 V to +1.2 V.

## RESULTS AND DISCUSSION

### Glancing Angle X-ray Results

The glancing angle x-ray diffraction patterns (GAXRD) for the undoped BSTO thin films deposited on MgO and LaAlO<sub>3</sub> are shown in Figures 1(a) and 1(b). Both the films were deposited under a oxygen partial pressure of 50 mT using the same ceramic ablation target. As shown in the figures, the GAXRD gives evidence of the existence of a secondary phase (Ba<sub>0.91</sub>Sr<sub>0.09</sub>TiO<sub>4</sub>) in the thin films whereas, the x-ray diffraction pattern of the target (inset) does not show the presence of this secondary phase.

In order to examine the role of the type of oxide bottom contact layer, the undoped thin films were also deposited on RuO<sub>2</sub>/sapphire substrates. Figures 2(a) and 2(b) show the GAXRD of undoped BSTO thin films deposited on RuO<sub>2</sub>/sapphire substrates under the oxygen partial pressures of 150 mT and 50 mT. The GAXRD does not indicate the presence of the same secondary phase (Ba<sub>0.91</sub>Sr<sub>0.09</sub>TiO<sub>4</sub>) when the film was deposited under a partial pressure of 150 mT on the RuO<sub>2</sub>/sapphire substrate but the secondary phase was present for the 50 mT sample. It is important to note that the same BSTO ceramic ablation target was used for all the thin film depositions shown in Figures 1(a), 1(b), 2(a), and 2(b).

## Electronic Measurements

Fig. 3 shows the capacitance versus voltage characteristics for the undoped BSTO film deposited on RuO<sub>2</sub>/Sapphire ( $P_{\text{oxygen}} = 150 \text{ mT}$ ) measured at 30 KHz. The dielectric constant at a zero bias was about 1470 with a tunability of about 40% at a field of  $2.0 \text{ V}/\mu\text{m}$ . The curve shows a symmetric capacitance-voltage relationship which is characteristic of paraelectric films. Figure 4 shows the C-V curve for the undoped BSTO film deposited on RuO<sub>2</sub>/sapphire deposited at a partial pressure of 50 mT also measured at 30 KHz. A dielectric constant of 1280 and a tunability of 48 % ( $V_{\text{applied}} = 2.0 \text{ V}/\mu\text{m}$ ) was obtained. A dielectric constant of 940 and a tunability of 55% ( $V_{\text{applied}} = 3.3 \text{ V}/\mu\text{m}$ ) were obtained for the same thin film at 0.5 MHz.

It is evident that the tunability of the film is not significantly altered at the two frequencies (30 KHz and 0.5 MHz) due to the presence of a secondary phase. Also, the dielectric constant and the tunability of the two BSTO films deposited under different oxygen partial pressures seem to be within acceptable range ( $< 15\%$ ) of each other. Also, any porosity and/or leakage current in the films will tend to alter the dielectric constants obtained. The bulk form of the undoped material has a dielectric constant of 3300 and a tunability of 20 % at  $0.73 \text{ V}/\mu\text{m}$ .

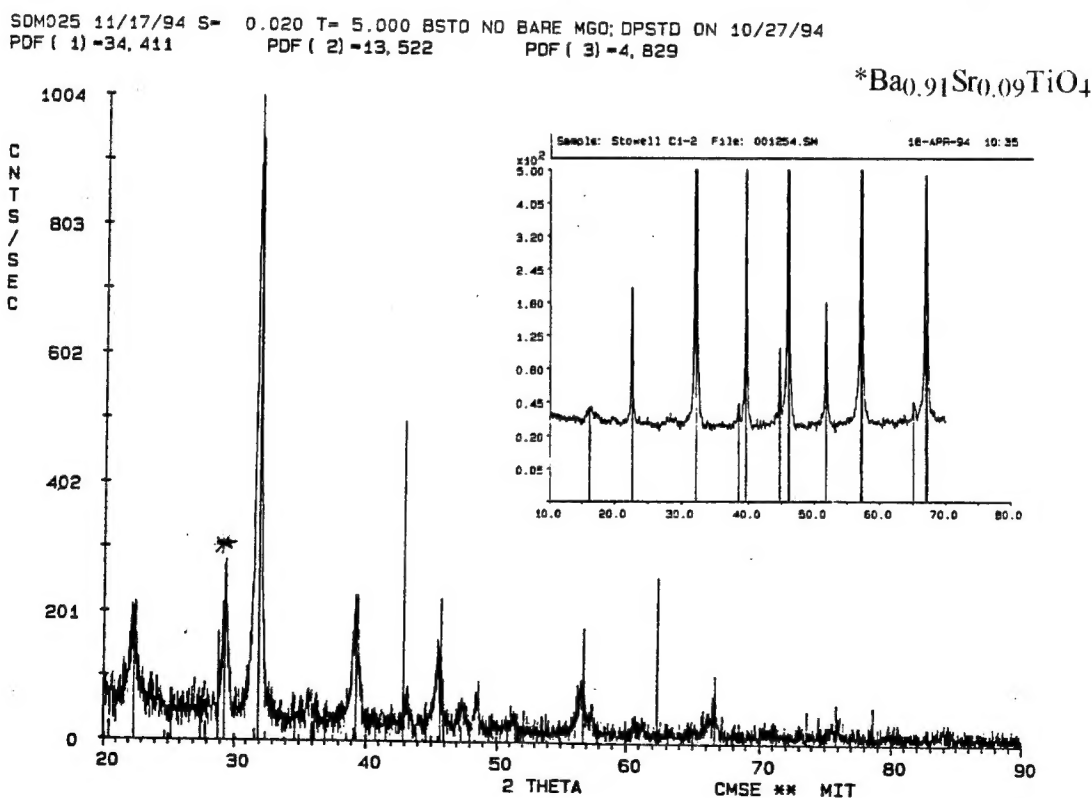


Fig. 1(a). Glancing angle x-ray diffraction pattern of undoped BSTO film deposited on MgO. Oxygen partial pressure = 50 mT. Inset shows x-ray pattern of the target material.

SOM026 11/17/94 S= 0.020 T= 5.000 BSTO ON BARE LAAL03; DPSTD ON 10/27/94  
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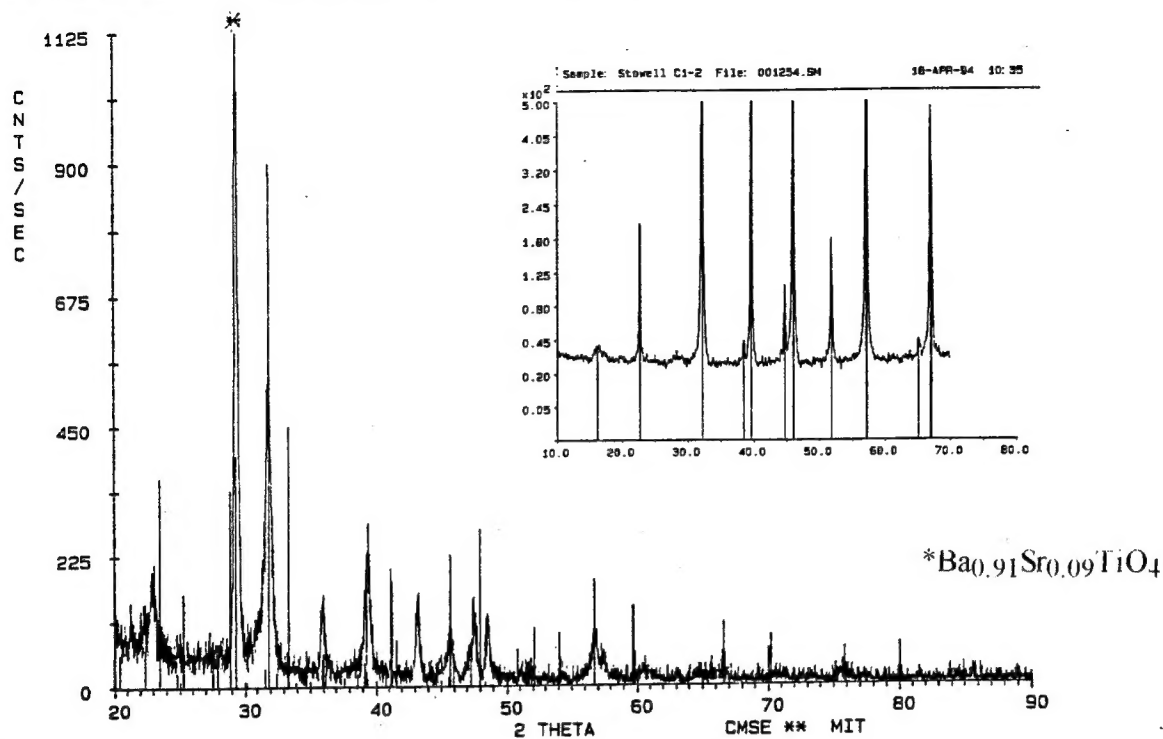


Fig. 1(b). Glancing angle x-ray diffraction pattern of undoped BSTO film deposited on LaAlO<sub>3</sub>. Oxygen partial pressure = 50 mT. Inset shows x-ray pattern of the target material.

SOM024 11/17/94 S= 0.020 T= 5.000 BSTO/RUO2/SAFIR; SAMPLE A  
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 PDF ( 5 ) =10, 173

PDF ( 4 ) =13, 522

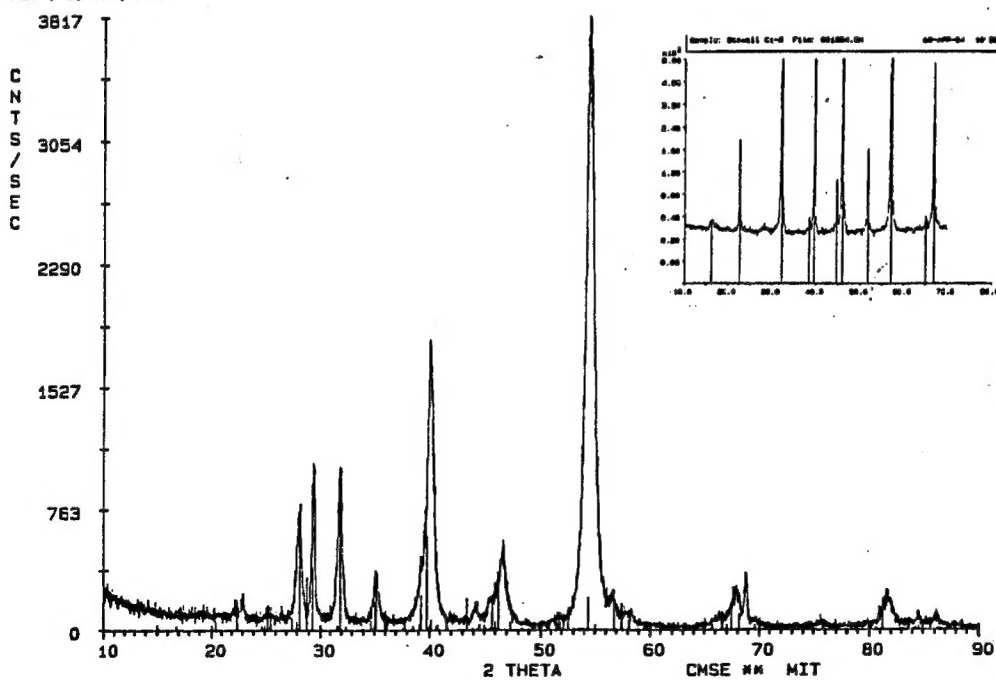


Fig. 2(a). Glancing angle x-ray diffraction pattern of undoped BSTO film deposited on RuO<sub>2</sub>/Sapphire. Oxygen partial pressure = 150 mT.



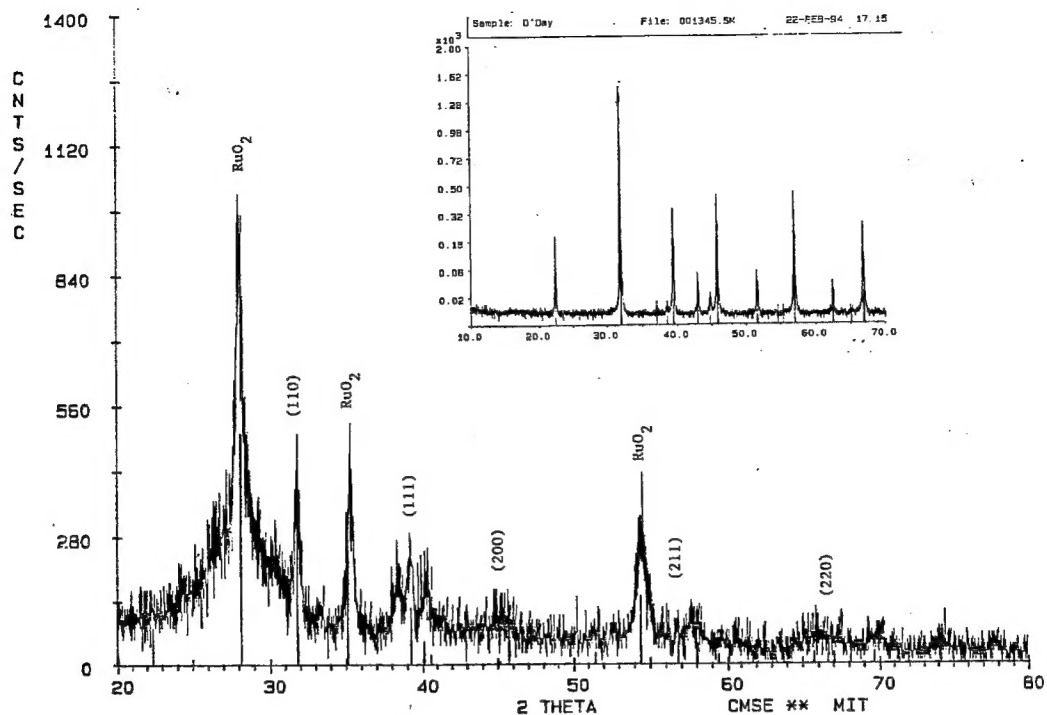


Fig. 2(b). Glancing Angle X-ray diffraction pattern of undoped BSTO film deposited on RuO<sub>2</sub>/Sapphire. Oxygen partial pressure = 50 mT. The broadening of the peaks are attributed to the inherent noise level of the GAXRD system.

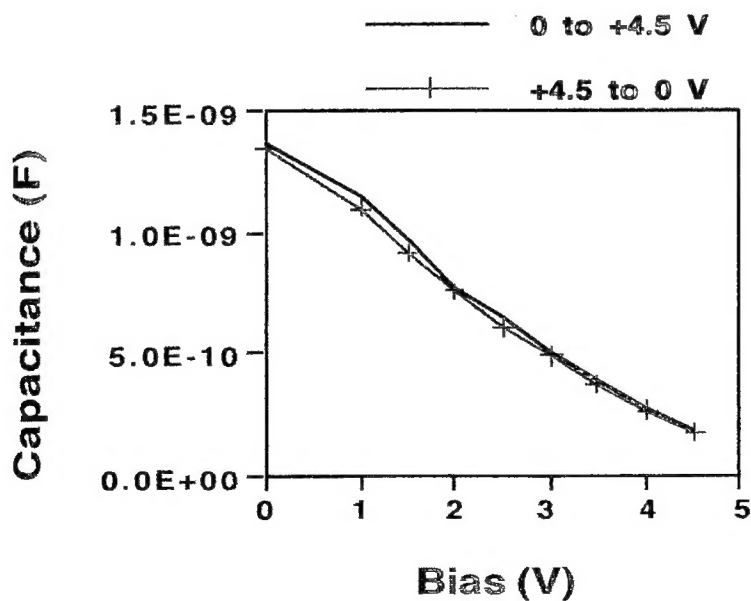


Fig. 3. Capacitance versus Voltage characteristics of BSTO thin film deposited on RuO<sub>2</sub>/Sapphire. P<sub>oxygen</sub> = 150 mT.

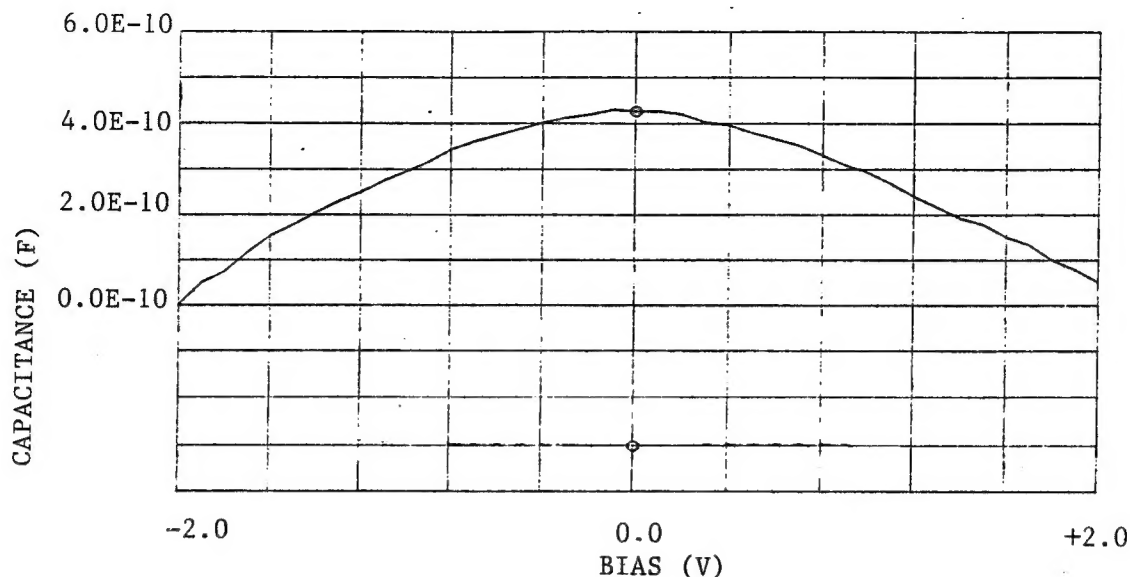


Fig. 4. Capacitance versus Voltage characteristics of the BSTO thin film deposited on RuO<sub>2</sub>/Sapphire. P<sub>oxygen</sub> = 50 mT.

### CONCLUSION

It has been demonstrated that the presence of a small amount of secondary phase in the BSTO thin film may not significantly alter the electrical characteristics of the thin film. Establishing the percentage of a secondary phase that may alter its performance is under investigation using relative peak height ratios from GAXRD studies. The ongoing study will establish the tolerance factor for the composition and processing variations of a BSTO thin film for a particular device application. Similar investigations for oxide modified BSTO thin films is also underway.

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